



Accelerator Mass Spectrometry & Ion Beam Analysis at ANSTO's

Centre for Accelerator Science

A.M. Smith (and a cast of thousands!)

Australian Nuclear Science and Technology Organisation (ANSTO), Locked Bag 2001, Kirrawee NSW 2232, Australia.

New User Symposium, Australian Synchrotron, 6th September 2016, National Centre for Synchrotron Science

overview

- What is the Centre for Accelerator Science (CAS)?
- What are *particle accelerators* and *how do they work?*
- What is 'Accelerator Mass Spectrometry' (AMS)?
- What is Ion Beam Analysis (IBA)?
- Example: ¹⁴C or 'radiocarbon'.

Centre for Accelerator Science at ANSTO



CAS project leader: Michael HOTCHKIS Project Manager: Jason COWAN & Shane HARRISON Construction Supervisor: Mark HARRIS Building construction: KANE PTY LTD Architects: DARYL JACKSON ROBIN DYKE PTY LTD National Electrostatic Corporation Accelerators: Eric ALDERSON, Alan OCONNOR, Richard KITCHEN, ...

Accelerator Operations: David BUTTON, Philip CHATFIELD, Andrew DOWNES, Peter DREWER, Oliver EVANS, David GARTON, Shaun KOZANIC, Shane LONG, Damien LYNCH, Michael LYNCH, Michael MANN, Tony MOWBRAY, Craig Robert THOMPSON, Jian WANG

Accelerator Mass Spectrometry Scientists: David CHILD, David FINK, Toshiyuki FUJIOKA, Michael HOTCHKIS, Quan HUA, Andrew JENKINSON, Vladimir LEVCHENKO, Andrew SMITH, Klaus WILCKEN, Bin YANG

Accelerator Mass Spectrometry chemists: Linda BARRY, Fiona BERTUCH, Brodie BISHOP, Geraldine JACOBSEN, Steven KOTEVSKI, Shwaron LAL, Charles MIFSUD, Tan NGUYEN, Prabha PRATAP, Krista SIMON, Simon SIMON, Simon VARLEY, Alan WILLIAMS

Ion Beam Analysis Scientists: Armand ATANACIO, David COHEN, Mihail IONESCU, Zeljko PASTUOVIC, Rainer SIEGELE, Ed STELCER ANSTO support staff & contractors: MANY!

Australian Nuclear Science and Technology Organisation Lucas Heights, Sydney

HIFAR reactor

Centre for Accelerator Science ANTARES 104 [B53]

OPAL reactor

(20 MW)

new accelerators

<mark>S c</mark>aemistr

Centre for Accelerator Science



CAS Research and Technology Platform

Isotopic ultra-tracing, nuclear forensics, bomb pulse, actinides, bio uptake, diet, toxicology Micro-spectroscopy, 2D mapping, Bio-imaging, Micro-implantation

Ion source enhancement, ECR ion source development, IRMS++

Environmental tracing, Hyper-accumulating plants systems, salinity, erosion, environmental sensors

Fine particle air pollution

Archaeology . Geology

Earth sciences, Geomorphology, landscape change, Climate change, Water management

CAS Platform

Facilities 4 accelerators: 1MV, 2MV, 6MV & 10MV AMS Sample Preparation Lab Low Energy Ion implanters

CapabilitiesAccelerator Mass Spectrometry (AMS)Ion Beam Analysis (IBA)Technical Group: maintenance & development

Users: universities (37), AINSE, CSIRO, IAEA, international collaborators, commercial, training Fundamental physics, nuclear reactions, X-ray cross sections, Subshell fluorescence yields, Heavy ion stopping, neutron cross sections, mono-energetic neutrons, Transmutation doping

> Radiation dosimetry, Micro-dosimetry, Micro-detectors, IBIC mechanisms

Materials research (modification & characterisation) Structural and Functional Interface engineering, Ion beam deposition, Thin films & coatings, Ion implantation, Ion damage, Structural Nuclear Materials

Centre for Accelerator Science: CAS

Three user groups:

- AMS (Accelerator Mass Spectrometry). AMS samples go into the ion source.
- IBA (Ion Beam Analysis). IBA samples go into the end stations.
- ASD (Accelerator Systems and Development) Maintenance and development.

CAS facilities

Four tandem accelerators:

- ANTARES: 10MV, HVEC, 1988, AMS & IBA
- STAR: 2MV, HVEE, 2003, AMS & IBA
- VEGA: 1MV, NEC, 2014, AMS
- SIRIUS: 6MV, NEC, 2015, AMS & IBA
- AMS Sample Preparation Facility
- Gas filled magnet (isobar separation)
- Ion implanters

🗯 🧠 👘 **Research Portal**

Welcome

to ANSTO's Interim Research Portal.

This is your gateway to all of ANSTO's research facilities and experience.

The 2017-1 Proposal Round for access to ANSTO's facilities and capabilities from January 2017 is now open. The deadline for proposals is 30 September 2016.

What is the ANSTO Interim Research Portal?

From 15 February 2016, this interim research portal will accept new proposals for access to facilities and capabilities at the Australian Nuclear Science and Technology Organisation (ANSTO), with the exclusion of the neutron-beam facilities at OPAL and the National Deuteration Facility.

A new ANSTO Research Portal will be available later this year and, for the first time, will provide one central location for the submission of proposals and subsequent experiments at ANSTO. The new ANSTO Research Portal is being designed to harmonise arrangements and processes across ANSTO to better support our user community.

ANSTO is one of Australia's largest public research organisations and custodian of much of our country's landmark and national research infrastructure, including the Open Pool Australian Lightwater (OPAL) multipurpose research reactor, the Australian Synchrotron, the Centre for Accelerator Science and neutron beam instruments.

On average, ANSTO accommodates over 1800 visiting researchers from other Australian research organisations and international research centres each year to provide access to a wide range of world-class research facilities that support research into human health, our environment and innovation for industry.

As outlined above, this new interim research portal has been put in place for the first round of proposals for 2016. Proposals related to environment, archaeology, geoscience, material science and engineering, life sciences, biomedical and human health should be submitted through this interim portal.

Simply use your email address to register an account above.

You can then use the portal to submit proposals, edit proposals, add experimenters/collaborators and more.

If you encounter an issue with using this interim portal or have any other questions or queries, please contact Joseph Bevitt via the user office by email or by phone on (+61) 2 9717 7232.

Further changes and improvements will be made to the portal in time for the following proposal round later in 2016.

If your proposal is not related to the above facilities and capabilities, or is specifically related to OPAL and the neutron beam facilities, please use the Bragg Institute Customer Portal.

Round 2017-1 for access 1 Jan'17 to 30 Jun'17 closes on 15 September 2016

Round 2017-2 for access 1 Jul'17 to 31 Dec'17 closes on 15 March 2017





Sign in

Password:

Login

ams@ansto.gov.au

Forgot your password?

Register

Email:

Bird building (B53)



Home of ANTARES, SIRIUS & VEGA accelerators.



Bird building accelerators



07 July 2015 11:41

ANTARES: 10MV (B53)



ANTARES: 10MV, HVEC, 1988, AMS & IBA

IBA beamlines:

Heavy ion microprobe Elastic recoil detection (ERDA) Neutron Ion irradiation

NTARES

AMS beamlines: ⁷Be, ¹⁴C, ²⁶Al, ³⁶Cl ¹⁰Be ¹²⁹I & Actinide Gas filled magnet

VEGA: 1MV, NEC, 2014, AMS





20. Degrader Foil

9. 90° Magnet

8.

- 10. Multi-Faraday Cup
- 11. Turbo Pumping Station, Slit,

Turbo Pumping Station

- Beam Profile Monitor (BPM) and Faraday cup
- 12. Einzel lens

- 23. 120° Magnet24. Multi-Faraday cup
- 25. Detector

21. 90° ESA

22. BPM

SIRIUS: 6MV, NEC, 2015, AMS & IBA

BA beamlines:

Confocal heavy ion. Surface engineering beamline. Nuclear reaction analysis & channelling beamline. Heavy ion implantation.

AMS beamlines: Time of flight AMS beamline. ¹⁰Be AMS beamline. ¹⁴C, ³⁶CI AMS beamline.



STAR: 2MV (B22)





IBA beamlines:

Multi elemental surface analysis High resolution depth profiling Ion Irradiation

AMS beamline: ¹⁴C (recombinator)

Libby building (B29)











Libby building: AMS chemistry

- Quarantine laboratory
- Ice core lab: -20° freezer
- Actinides & iodine laboratory
- Speleothem laboratory
- Tree ring laboratory
- Multi-purpose laboratory
- Cosmogenic laboratory (rock crushing & sieving, heavy liquid & Franz separation, ultrasonic & acid)
- ¹⁴C in situ dating laboratory
- Stable isotope laboratory



Stage 2: relocation of ¹⁴C chemistry from B16.

What is a particle accelerator?

Particle accelerators are tools that produce 'beams' of charged particles (electrons, atoms or molecules) for identification or to study physical phenomena or interaction.



Electric Field

Magnetic Field (Field coming out of the page)







Accelerator Charging Systems

Pelletron charging system







Diagram: http://www.pelletron.com/charging.htm



Tandetron solid-state charging system





insulating gas: sulphur hexafluoride (SF₆)



GWP 22,800 over 100 years!

Accelerator beam optical elements

- Acceleration: acceleration tubes, gap lenses
- Mass selection: double-focusing dipole magnets
- Energy & charge selection: electrostatic analysers (cylindrical & spherical)
- Velocity selection: E×B filters (Wien filter)
- Focusing: Einzel lenses, quadrupoles (magnetic & electrostatic, doublets & triplets)
- Steering: electrostatic & magnetic deflectors
- Beam definition: slits & apertures
- Beam shape: beam profile monitors
- Vacuum pumps: need high vacuum (~ 10⁻⁶ Pa) to avoid collisions with background gas (causes charge and trajectory change)

What is Accelerator Mass Spectrometry?

- AMS (Accelerator Mass Spectrometry). AMS samples go into the ion source.
- An ultra-sensitive method for measuring isotopic ratios.
- AMS differs from other forms of mass spectrometry in that it accelerates ions to high kinetic energies before mass analysis.
- AMS is based on <u>atom counting</u> so it is inherently sensitive.
- AMS is typically used for isotopes that have intermediate halflives and/or very low abundance: *often 'cosmogenic'*.
- Thus AMS finds many applications in Earth Sciences, archaeology, biomedicine, nuclear safeguards and nuclear forensics, to name but a few!



ANSTO AMS radionuclides

radioisotope half-life (years)

'cosmogenic'						
⁷ Be 53.28 days						
¹⁰ Be	$1.39 imes 10^{6}$					
¹⁴ C	5,730					
²⁶ AI	$7.3 imes10^5$					
³⁶ Cl	$3.01 imes10^5$					
129	$15.9 imes10^6$					
'primordial'						
²³⁵ U 7.04 × 10 ⁸						
236 U	2.342×10^7					
'anthropogenic'						
²³⁹ Pu	2.410×10^4					
²⁴⁰ Pu	$6.56 imes10^3$					
42Pu (as 'spike')	$3.75 imes 10^5$					



AMS isotopes & applications

Common AMS isotopes (half-lives):

carbon-14 (5730 a), beryllium-10 (1.39 Ma), chlorine-36 (301 ka), aluminium-26 (720 ka), iodine-129 (15 Ma), calcium-41 (103 ka), actinides (various)

• Applications (incomplete list!):

archaeology, exposure-age dating, landscape evolution & catastrophic events, global climate change, pollution, biomedicine, oceanography, hydrology, extra-terrestrial material, nuclear physics, nuclear safeguards, materials analysis,

• Other AMS isotopes (half-lives, applications):

tritium (12.5 a, biomed), beryllium-7 (53 days, tracing), silicon-32 (~160a, dating, biomed), manganese-53 (3.8 Ma, meteorites, supernova, biomed, nuclear waste), iron-60 (1.5 Ma, supernova), nickel-63 (100 a, fast neutron dosimetry), selenium-79 (1.1 Ma, biomed), strongtium-90 (28.5 a, nuclear safeguards)

Principle of radiocarbon dating



- ¹⁴C or radiocarbon produced by neutron capture on ¹⁴N.
- Photosynthesis: living organisms are in equilibrium with the atmosphere.
- The radiocarbon clock starts on death: ¹⁴C uptake ceases.
- Radiocarbon dating method: Willard F. Libby, Chicago 1950.
- Initially *Radiometric* and then *accelerator mass spectrometry*.



credit: M. Blaauw 2007, chrono.qub.ac.uk/blaauw

- Black square is carbon, mostly ¹²C (99%) and ¹³C (1%).
- Yellow dots are ¹⁴C atoms, initially 10⁴ atoms.
- ¹⁴C atoms are radioactive and disintegrate with a half-life of 5,730 years.
- When? It cannot be predicted for a given atom.
- Dating old samples is difficult: few ¹⁴C atoms remain.
- Modern natural carbon contains ~ 50 million per mg.

How do we actually do ¹⁴C AMS?

- Aim: to determine the ¹⁴C/¹²C ratio ('activity') for a sample.
- The sample must firstly be physically and chemically purified to isolate the carbon: *this can be complex & time consuming!*
- The sample carbon is ionised in a negative ion source and beams of ¹²C⁻, ¹³C⁻ and ¹⁴C⁻ are produced.
- These beams are accelerated in a tandem accelerator and are stripped to positive beams at the central stripper then further accelerated.
- ANTARES: terminal voltage 5.2 MV, ¹⁴C⁴⁺ @ 26 MeV, isotope bouncing
- STAR: terminal voltage 2 MV, ¹⁴C³⁺ @ 8 MeV, recombinator

detection

- Stable ¹²C and ¹³C beams are measured by charge digitisation.
- Rare radioactive ¹⁴C events counted in an ionisation detector.
- Unknowns are measured against ¹⁴C standards and ¹⁴Cdepleted material.



Why use tandem AMS ¹⁴C measurement?

negative ions: elimination of ¹⁴N isobar charge exchange: destruction of ¹²CH₂ & ¹³CH in terminal ionisation detector: E,M,Z \Rightarrow atom counting!

features:

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ultra-small samples ~ 0.1 mg: at ANSTO ~5µg
rapid measurement ~ 20 min
sensitivity: 1 in 10<sup>16</sup>: atom counting!
accuracy ~ 0.3%, background ~ 50 ka
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Micro-sample (~5-100µgC) applications

climate change

- CO, CO₂ & CH₄ in ice sheets [major driver]
- particulate matter in ice sheets
- aerosol fractions for source discrimination/apportionment: elemental carbon, organic tracers & toxic compounds

chronology, archaeometry

- Palynomorphs: pollen, microforamifera, phytoliths, diatoms
- organic fractions from archaeological material (mummies, cooking vessels, residues...)

biomedical

DNA (eg for brain/neuron cell age), specific compounds
 Preparative gas chromatography

CO, CO₂ & CH₄ from polar ice sheets



















Sources of methane

Natural processes













Wetlands

Termites

Wild animals

Oceans

Geologic

Wildfires

Human activity



Rice cultivation











Landfills

Wastewater

Livestock

Gas & petroleum Coal mining

Identifying origin of methane



Palynomorphs



Pollen from flowering plants and a cone-bearing plant (pine). [All Scale bars = $10\mu m$.]



L-R: foramifera, phytoliths, diatoms.

artefact wear & use residues



Radiocarbon 'dating' human cortical neurons

How old are the neurons in the cerebral cortex of the human brain? ANSTO collaboration with the Medical Nobel and Karolinska Institutes, Sweden

- ANSTO's international reputation for *microgram* ^{14}C dating DNA samples at 10 μ g
- Exploits atmospheric doubling of ¹⁴C due to nuclear weapons testing in the 60's



Adult cortical neurons have ¹⁴C levels corresponding to time of birth suggesting an absence of cortical neurogenesis

Neocortical neurogenesis in humans is restricted to development

Ratan D. Bhardwaj*[†], Maurice A. Curtis[†]⁺, Kirsty L. Spalding⁺, Bruce A. Buchholz⁶, David Fink¹, Thomas Björk-Eriksson¹, Claes Nordborg⁺⁺, Fred H. Gage⁺⁺, Henrik Druld⁺⁺, Peter S. Eriksson⁺⁵⁵, and Jonas Frisén⁺⁵⁵

12564-12568 | PHAS | August 15, 2006 | vol. 103 | no. 33

NEUROSCIENCE

No More Cortical Neurons for You

Pasko Rakic

18 AUGUST 2006 VOL 313 SCIENCE www.sciencemag.org

What is Ion Beam Analysis?

- IBA (ion beam analysis).
 IBA samples go into the end stations.
- Ion Beam Irradiation: Chemistry, Defects, Damage, Neutron Production
- Ion Beam Characterization:

Rutherford Backscattering Spectroscopy (RBS, C-RBS); Particle Induced X-ray Emission (PIXE); Elastic Recoil Detection Analysis (ERDA); Nuclear Reaction Analysis (NRA); Heavy Ion Micro-probe (HIMP).

IBA Characterisation Techniques

Analytical Technique	Typical Applications	Elements Detected	Sensitivity	Depth Resolution	Analysis Depth
<u>RBS</u>	Surface and thin film composition and thickness.	Li - U	Best for heavy elements on light element substrate (eg Cu on Si) typical 10% (Li) - 0.001% (U)	5-20 nm	up to 1 mm
<u>PIXE</u>	Trace element composition of particulates and bulk materials.	Si-U	Optimum near Fe (1ppm) elemental sensitivities range from 1 - 100ppm	typical proton range 20- 50 microns	up to 1 mm
<u>PIGE</u>	Trace light element composition of particulates and bulk materials.	Li-Al	Element dependent, typical < 1 ppm for F < 40 ppm for Na < 40 ppm for Al	-	up to 1 mm
<u>NRA</u>	Isotopic tracing and profiling in materials, surfaces and interfaces.	H-Si	Element dependent, typically in range 1 - 100 ppm	5-20 nm	up to 1 mm
<u>PESA</u>	Hydrogen in thin polymers; polymer interdiffusion, hydrogen in solar cells.	H, D	> 0.1%	10-20 nm	up to 1 mm
ERD & <u>RToF</u>	Elemental composition and structure of near surface regions, interfaces, thin films.	H-U	> 0.1 %	10-20 nm	up to 1 mm
<u>µPIXE</u>	Trace element mapping in biological, environmental and geological samples.	Si-U	> 100 ppm	-	up to 3 μm
<u>µERDA</u>	Elemental mapping and depth profiling of hard surfaces.	H-U	> 0.1 %	> 50 nm	up to 20 μm
<u>IBIC</u>	Charge collection mapping in electronic devices and detectors.				up to 1 µm



$$E_{0x} = E_0 - \frac{x}{\cos \alpha} \frac{dE_0}{dx} \qquad N[at/cm^2] = \frac{Y[cts]\cos \alpha}{N_i \Omega \frac{d\sigma}{d\Omega}} \qquad N_i - \text{number of ions incident}$$

$$E_{1x} = kE_{0x} \qquad N[at/cm^2] = \frac{Y[cts]\cos \alpha}{N_i \Omega \frac{d\sigma}{d\Omega}} \qquad \Omega_i - \text{detector solid angle}$$

$$E_2 = E_{1x} - \frac{x}{\cos \beta} \frac{dE_{1x}}{dx} \qquad \frac{d\sigma}{d\Omega} = \frac{[Z_1 Z_2 e^2 (M_1 + M_2)]^2}{4E_0^2 M_2^2 \cos \theta} \qquad \sigma - \text{scattering cross section}$$

nt on sample n

Rutherford Backscattering

Applications:

- Elements heavier than the projectile •
- Mostly suitable for heavy elements in a light elements matrix
- Depth of analysis: few nm up to few µm
- Depth resolution: few 10nm •
- Sensitivity: few at% for light elements; few 100ppm for heavy elements



RBS Example



RBS spectrum (θ = 170°) for 3.0 MeV He⁺ ions incident on a 400 nm Al film with thin Au markers on the front and back surfaces

Proton Induced X-Ray Emission (PIXE)

Applications: Trace element (AI to U) composition of particulates and bulk materials



- Optimum sensitivity near Fe (1ppm)
- Elemental sensitivities range 1-100 ppm
- Typical proton depth range 20-50 microns

Proton Induced Gamma Emission (PIGE)

Applications:

• Trace analysis of light elements (Li to AI) in particulates, bulk materials and thin films

Sensitivity element dependent, typical < 1 ppm for F; < 40 ppm for AI and Na



Nuclear Reaction Analysis (NRA)

Applications:

- Depth profiling of light elements: H-Si
- Depth of analysis: few nm to few μm
- Depth resolution: 5-20nm
- Sensitivity: 1-100ppm
- Applicable for crystalline & amorphous

Target Element	Input Beam	Resonance Energy [keV]	Max. Energy [keV]	Input Beam Current	Max. TV [MV]	E _y [MeV]
⁷ Li	$^{1}\mathrm{H}$	2,060	5,000	500nA	2.5	16.15
¹⁰ B	⁴ He	2,400	5,000	400nA	2.5	3.8
¹⁶ O	⁴ He	3,040	6,000	400nA	3	1.6
¹ H	^{15}N	6,385	15,000	200nA	4	4.43





- Thick DLC film grown by DC-PACVD on Si(100) using CH₄ and C₂H₂ to vary Hydrogen
- Hydrogen content plays a role in the biologic response
- Hydrogen content is higher at the surface and decreases toward the interface



NRA example: Oxidation kinetics of Ta

Sample: Ta₂¹⁶O₅ film 200nm thick on polycrystalline Ta exposed to ¹⁸O Result: ¹⁸O depth profile





Elastic Recoil Detection Analysis (ERDA)



Applications:

- Elements: lighter than the projectile
- Depth of analysis: up to few 100nm
- Depth resolution: few 10nm
- Sensitivity: 0.1 at% to few at%

There are three options for the type of energy detector:

- SB detector
- dE-E detector
- Time-of-flight detector

ERDA example:82 MeV I and ToF detector

Sample: LiNbO/Si(100) Results: Film thickness ~100ML ~6ML SiO₂ present at the interface Nb diffusion in Si

Quasi Mono-energetic neutrons

⁷Li(p,n)⁷Be Max E p = 20 MeV Max E n = 18 MaV

Applications: Isotope production, study of nuclear reactions, study of nuclear materials, etc.

- Number of n produced: $dN = ig\rho_{Li} \frac{d\sigma}{d\Omega} E_p dx d\Omega$
- Where: *i* ion beam current [μ A] *g* –no of protons/ μ A ρ_{Li} –density of ⁷Li $d\sigma/d\Omega$ -differential cross section in the lab frame E_p -energy of protons

- Energy of n:

$$E_{n} = \frac{m_{p}m_{n}}{(m_{Li} + m_{n})^{2}} [\mu \pm \xi]^{2} E_{p}$$

Where: $\mu = \cos\theta$; θ -n direction in the lab frame;

$$\xi^{2} = \sqrt{\frac{m_{Li}(m_{Li} + m_{n} - m_{p})(E_{p} - E_{th})}{m_{p}m_{n}E_{p}}} + \mu^{2} - 1$$

 E_{th} =1.881MeV for ground state reaction E_{th} =2.378MeV for excited state reaction

Thanks for your interest!

Ginsto

Nuclear-based science benefiting all Australians

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